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FERROMAGNETIC RELAXATION IN ULTRATHIN FILMS OF  
AMORPHOUS ALLOYS(U) JOHNS HOPKINS UNIV BALTIMORE MD  
DEPT OF PHYSICS AND ASTRONOMY G T RADO 84 APR 86

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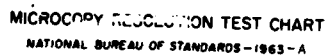
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FINAL TECHNICAL REPORT

on the research project

Ferromagnetic Relaxation in Ultrathin Films of Amorphous Alloys

funded by Naval Research Laboratory contract

N00014-85-K-2027

Submitted by:

George T. Rado, Principal Investigator  
Research Professor of Physics  
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## INTRODUCTION

This document is the final report on the research project "Ferromagnetic Relaxation in Ultrathin Films of Amorphous Alloys" which was funded by NRL contract N00014-85-K-2027 for the seven month period 15 July 1985 to 15 February 1986. A copy of the March, 1985 proposal for the present research and a copy of the NRL statement of work are appended for ease of reference.

We begin by reviewing the motivation of our work and its significance for the problem of electromagnetic pulse (EMP) mitigation. This is followed by a brief account of our scientific results. A more detailed presentation of these results is contained in the attached reprint of a scientific paper entitled "Magnetic surface anisotropy of amorphous Fe-B ultrathin films" by L. Zhang, G. T. Rado, S. H. Liou and C. L. Chien. This paper was presented orally by the Principal Investigator (P.I.) at the 1985 International Conference on Magnetism in San Francisco and was accepted for publication in the Journal of Magnetism and Magnetic Materials. The results of this paper appear also in a dissertation submitted by L. Zhang to The Johns Hopkins University in conformity with the requirements for the Ph.D. degree.

As stated in the March, 1985 proposal for the present research project, the use of ferromagnetic rather than nonferromagnetic materials offers several advantages in devices designed for EMP mitigation. It was suggested, moreover, that the shielding effectiveness of such devices might be further improved if their ferromagnetic compounds were made of amorphous rather than crystalline materials. In addition to being highly flexible, certain amorphous ferromagnets have an unusually low magnetostriction so that

the dependence of their magnetic properties on external stresses is relatively weak. Since the shielding effectiveness of a ferromagnetic material is not determined solely by its conductive properties, the present research involves investigations by means of ferromagnetic resonance (FMR) of the material's magnetic properties.

In accordance with the plans discussed in our proposal of March, 1985, the amorphous materials we used were in the form of ultrathin films. This choice offered new opportunities such as the first reliable measurements of magnetic surface anisotropy (which we accomplished successfully, as reported below) and the preliminary observation of a film-thickness-dependent FMR linewidth in ultrathin films. It should be recalled, in this connection, that FMR provides information on resonance fields (or frequencies) and also on linewidths, and that the latter are related to ferromagnetic relaxation, i.e., to magnetic losses. One cannot, however, successfully investigate relaxation phenomena unless one first understands the resonance fields, and this, in turn, necessitates measuring the magnetic surface anisotropy. An understanding of the resonance fields and a reliable measurement of the surface anisotropy are, in fact, the central contents of the present report. In regard to the NRL statement of work (appended to this report) for our seven month research program, we are pleased that while item 3.3 is in a preliminary stage, all the other items of that lengthy statement of work have been accomplished successfully.

If, as we hope, this research project continues to be funded after February 15, 1986, then we shall concentrate on measurements of relaxation in ultrathin amorphous films and on investigations of various structures consisting of several alternating layers of ultrathin amorphous ferromagnets

and conductive nonferromagnetic metals. With this flexible arrangement we should be able to control independently the magnetic and conductive properties of layered structures and to make use of the relaxation properties of ultra-thin films for achieving a sufficiently large total shielding in a relatively small space.

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## ACCOMPLISHMENTS

For reasons noted in the Introduction we focused our seven month research effort on developing a reliable and rigorous method for measuring the surface anisotropy of amorphous media and on applying that method to amorphous Fe-B alloys. Our method includes FMR experiments in several ultrathin ( $\leq 10^2 \text{ \AA}$ ) amorphous films and some novel theoretical interpretations. The latter involve an adaptation to amorphous films of a theory (see reference 1 of the attached reprint) proposed by the P. I. for crystalline films. We believe that previous methods for measuring the surface anisotropy are unsatisfactory: The "critical angle method" involves the unjustified assumptions that the static magnetization is necessarily quasi-aligned with the static magnetic field and that the mode of motion of the dynamic magnetization is generally circular rather than elliptical; the "dimensional spin wave resonance method", on the other hand, involves ambiguities in the order numbers of the spin wave peaks.

The essence of the above-mentioned theory for interpreting our FMR experiments on amorphous films is an appropriate solution of the Landau-Lifshitz equation of motion of the magnetization subject to the Rado-Weertman boundary conditions. (Literature references are given in reference 1 of the attached preprint.) We obtained this solution by requiring that our amorphous films satisfy the following assumptions: (a) the surface anisotropy energy density has the form  $-K_s \cos^2 \theta$ , where  $K_s$  is the surface anisotropy coefficient and  $\theta$  is the angle between the magnetization and the normal to the amorphous film; (b) the volume anisotropy energy density is zero; (c) the magnetization is homogeneous at the film surface as well as throughout

the film volume; (d) the film thickness is so small that the microwave magnetic field is uniform inside the film and dimensional spin wave resonances are absent. Because of the good agreement between our theoretical and experimental FMR results (see below), we believe that our ultrathin films, which were prepared by D.C. sputtering, do satisfy all the above-mentioned assumptions. It is particularly gratifying that the theory involves no approximations beyond the usual linearization of the equation of motion.

The formulas for the magnetic resonance fields  $H_{\text{res}}^{\parallel}$  and  $H_{\text{res}}^{\perp}$  derived by us are presented on page 5 of the attached reprint. These fields correspond, respectively, to the parallel FMR configuration ( $H \parallel \text{film}$ ) and to the perpendicular FMR configuration ( $H \perp \text{film}$ ). We find that for our Fe-B films the experimentally observed values of  $H_{\text{res}}^{\parallel}$  are larger than the values of  $H_{\text{res}}^{\parallel}$  calculated for the uniform mode. This means that the wave-numbers  $k_1$  and  $k_3$  are real, the modes are surface-induced, and the value of  $K_s$  is positive. In contrast, the experimentally observed values of  $H_{\text{res}}^{\perp}$  are found to be smaller than the values of  $H_{\text{res}}^{\perp}$  calculated for the uniform mode. The meaning of this result is that the wavenumber  $k$  is purely imaginary, the modes are spin waves (although their wavelengths are too long to permit dimensional resonances), and the value of  $K_s$  is positive, in agreement with our result for the parallel FMR configuration. A positive value of  $K_s$  signifies that at the film surfaces the easy axis of magnetization is perpendicular to the film.

Figure 1 of the attached reprint shows our experimental and theoretical FMR results at room temperature for the amorphous alloy  $\text{Fe}_{50}\text{B}_{50}$ . It is seen that good agreement obtains between the measured and calculated dependences on film thickness of  $H_{\text{res}}^{\parallel}$  at X-band,  $H_{\text{res}}^{\parallel}$  at K-band and  $H_{\text{res}}^{\perp}$

at X-band. The highest magnetic field at our disposal was only 13 kOe and thus too low for measurements of  $H_{\text{res}}^1$  at K-band. To determine the values of  $K_s$  and the magnetization  $M$  we fitted the theoretical curves to the experimental points. Particularly important is the fact that for each of the theoretical curves we obtained the same values of  $K_s$  and  $M$  (given in the figure caption) even though these curves involve two frequencies and two FMR configurations. Since the value of  $M$  agrees reasonably well with values found in the literature, it is seen that with our method we need to determine  $K_s$  only, i.e., just one unknown parameter.

Figure 2 of the attached reprint shows our experimental and theoretical FMR results at room temperature for the amorphous alloy  $\text{Fe}_{70}\text{B}_{30}$ . The measured and calculated dependences on film thickness of  $H_{\text{res}}^{||}$  are in good agreement at X-band and at K-band, just as in the case of  $\text{Fe}_{50}\text{B}_{50}$ . As to  $H_{\text{res}}^1$ , the above-mentioned unavailability of magnetic fields above 13 kOe prevented us from measuring  $H_{\text{res}}^1$  at either X-band or K-band. It should be noted, however, that from each of the two theoretical curves we again obtained the same values of  $K_s$  and  $M$  even though these curves involve two frequencies.

### CONCLUSION

On the basis of the information presented above and in the attached reprint (see, especially, Figs. 1 and 2), we believe to have accomplished the items listed on the NRL statement of work and also an understanding of the resonance fields, the development of a reliable and rigorous method for measuring the magnetic surface anisotropy of amorphous media and an application of that method to amorphous Fe-B alloys.

## MAGNETIC SURFACE ANISOTROPY OF AMORPHOUS Fe-B ULTRATHIN FILMS

L. ZHANG, G.T. RADO, S.H. LIOU and C.L. CHIEN

*Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, MD 21218, USA*

The magnetic surface anisotropy constants  $K_s$  of amorphous  $\text{Fe}_x\text{B}_{100-x}$  was determined by performing ferromagnetic resonance (FMR) measurements on ultrathin films and adapting a recent FMR theory to amorphous materials. For a given Fe content  $x$  the same value of  $K_s$  is obtained at two frequencies and two FMR configurations.

This paper reports on ferromagnetic resonance (FMR) measurements of the magnetic surface anisotropy energy of amorphous iron-boron alloys in the form of "ultrathin" films (thickness  $\leq 10^2$  Å). The method used in interpreting the measurements was developed by us specifically for ultrathin amorphous films. It includes an adaptation to amorphous materials of a recent theory [1] which treats the effects of surface anisotropy on the FMR in ultrathin monocrystalline films. This makes it possible to deduce the surface anisotropy constant  $K_s$  of an amorphous material from the dependence of the magnetic resonance field  $H_{\text{res}}$  on the film thickness  $2L$ . We find both theoretically and experimentally that under certain conditions the dependence of  $H_{\text{res}}$  on  $1/(2L)$  is linear. More importantly, we find that in ultrathin films our observed FMR modes are surface-induced modes [1] in the parallel FMR configuration ( $H \parallel$  film plane) and spin wave modes [but not the dimensional spin wave resonances (SWR) which are determined by the film thickness] in the perpendicular FMR configuration ( $H \perp$  film plane). This situation differs from that obtaining in the dimensional SWR experiments [2] customarily used for measuring  $K_s$ . The dimensional SWR experiments involve films which are "thin" (thickness  $\approx 10^4$  Å) rather than ultrathin and consequently their surface-induced and spin wave modes sometimes overlap. We also note that volume inhomogeneities of the magnetization may simulate surface anisotropies, and that it is probably easier to avoid such inhomogeneities in an amorphous film which is ultrathin rather than merely thin. Our experimental data are, in fact, reproducible within experimental error if the film preparation conditions are kept constant.

The samples for our measurements were prepared at room temperature by sputtering amorphous films of  $\text{Fe}_x\text{B}_{100-x}$  alloys (where  $x$  denotes atomic percent) at a rate of about 1 Å/min in a "sandwich" configuration between two 170 Å thick aluminum films. Glass substrates of 0.12 mm thickness were used. The sputtering current (consisting of highly pure ionized argon) was 0.25 A, and the pressure in the vacuum chamber prior to sputtering was typically  $2 \times 10^{-7}$  Torr. None of the films was annealed but the sputtering targets were an-

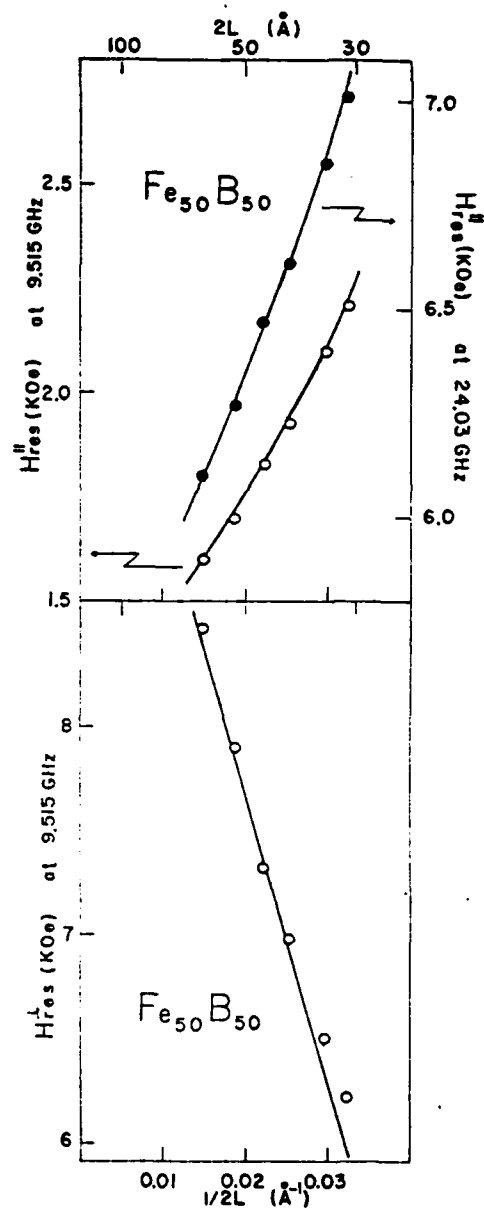


Fig. 1. FMR in  $\text{Fe}_{50}\text{B}_{50}$ : Experiment (data points) and theory (lines based on  $K_s = 0.20$  erg/cm<sup>2</sup>,  $M = 570$  emu, and a weak dependence on  $A = 5.5 \times 10^{-7}$  erg/cm).

nealed for 2 h at 600°C in an argon atmosphere. Films having  $x = 50$  and  $x = 70$  were prepared with thicknesses ranging from 70 to 30 Å and from 50 to 18 Å, respectively. The film thicknesses were determined by assuming that they are proportional to the sputtering times and then sputtering relatively thick ( $\approx 10^3$  Å) calibration films onto polished silicon wafer substrates. The thicknesses of these calibration films were measured to about  $\pm 10\%$  with a Sloan Dektak II surface profile measuring system. Some of the film thicknesses (not shown on the figure) were checked by X-ray fluorescence.

The FMR measurements were performed by means of standard microwave methods at room temperature. Magnetic fields up to 13 kOe were available. The films were mounted on teflon rods and inserted into the center of a rectangular TE<sub>102</sub> X-band cavity or a cylindrical TE<sub>011</sub> K-band cavity. For the  $x = 50$  films the parallel ( $\parallel$ ) FMR configuration was used at 9.52 GHz and at 24.03 GHz, and the perpendicular ( $\perp$ ) FMR configuration was used at 9.52 GHz. The experimental points and theoretical lines for the  $\parallel$  configuration are shown in the upper part of fig. 1 and for the  $\perp$  configuration in the lower part of fig. 1. We note that both experimentally and theoretically the sign of the slope of  $H_{res}$  vs.  $1/(2L)$  is opposite in the two configurations. For the  $x = 70$  films the  $\parallel$  configuration only was used at both 9.52 and 24.03 GHz, and the experimental points and theoretical curves (which are nonlinear for these films) are shown in fig. 2.

The theoretical curves are based on formulas which we derived for amorphous films in a manner analogous to that used in reference [1] for monocrystalline films. For the  $\parallel$  configuration we obtain

$$H_{res}^{\parallel} = -\frac{\pi}{2} M_{(-)} \left[ (2\pi M)^2 + (\omega/\gamma)^2 \right]^{1/2} + (2A/M) k_{1,3}^2,$$

with

$$2A(1 + \Delta) k_1 k_3 \tanh(k_1 L) \tanh(k_3 L) = K_s [(2 + \Delta) k_1 \tanh(k_1 L) + \Delta k_3 \tanh(k_3 L)],$$

and  $\Delta = \{1 + [\omega/(2\pi M\gamma)]^2\}^{1/2} - 1$ , where our experiments require that  $k_{1,3}$  be real. Here  $A$  is the exchange stiffness constant,  $\omega$  is the circular frequency and  $\gamma$  is the magnetomechanical ratio ( $g/2$ )  $2\pi(2.80)$  MHz/Oe.

For the  $\perp$  configuration we obtain

$$H_{res}^{\perp} = \omega/\gamma + 4\pi M + 2Ak^2/M,$$

$$\text{with } Ak \tanh(kL) = -K_s,$$

where our experiments require that  $k$  be purely imaginary.

In deriving these formulas we introduced neither a deposition-induced volume anisotropy nor an inhomogeneity in the magnetization  $M$  near the film surface. We assumed, however, that the microwave magnetic

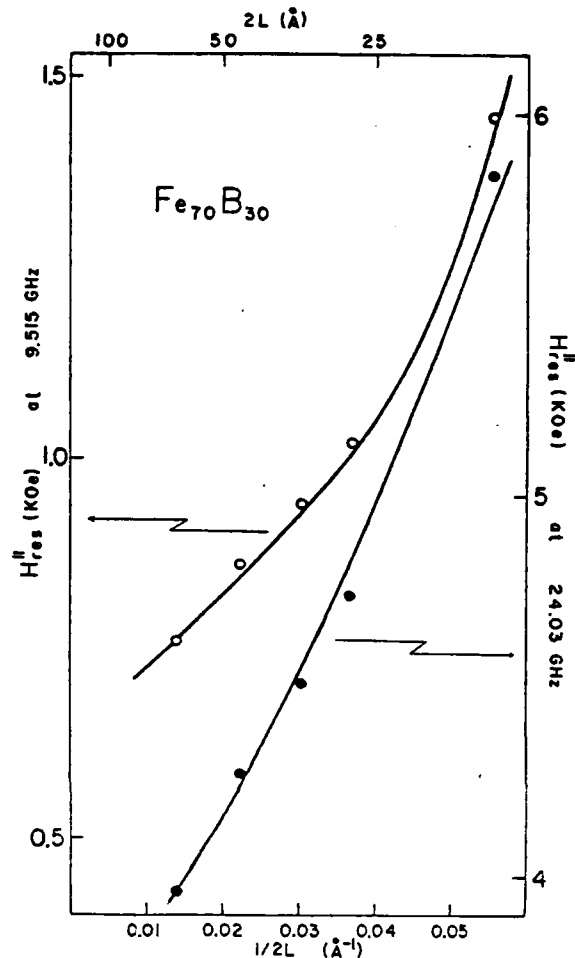


Fig. 2. FMR in Fe<sub>70</sub>B<sub>30</sub>: Experiment (data points) and theory (lines based on  $K_s = 0.53$  erg/cm<sup>2</sup>,  $M = 1240$  emu, and a weak dependence on  $A = 1.38 \times 10^{-6}$  erg/cm).

field in the film is spatially uniform. The cubic volume anisotropy is, of course, taken to be zero, and the surface anisotropy energy density  $E_{surf}$  of ref. [1] is replaced by  $E_{surf} = -K_s \cos^2 \theta$ , where  $\theta$  is the angle between  $M$  and the normal to the film plane. In our films  $K_s$  is found to be positive so that  $E_{surf}$  is of the easy axis type. We further note that our derivation involves no approximation beyond the usual linearization of the equation of motion. To determine the parameters  $K_s$  and  $M$  for a given  $x$  we used  $g = 2.09$  and fitted the theoretical curves to the experimental points. The good agreement between theory and experiment shown by figs. 1 and 2 is especially gratifying because for a given Fe content  $x$  the same  $K_s$  (and  $M$ ) is obtained at two frequencies and two FMR configurations.

[1] G.T. Rado, Phys. Rev. B26 (1982) 295, and erratum in Phys. Rev. B32 (1985) 6061.

[2] See, for example, L.J. Maksymowicz and D. Sendorek, J. Magn. Magn. Mat. 37 (1983) 177, and references contained therein.

PROPOSAL

TO: NAVAL RESEARCH LABORATORY

FROM: THE JOHNS HOPKINS UNIVERSITY  
DEPARTMENT OF PHYSICS AND ASTRONOMY  
CHARLES AND 34TH STREETS  
BALTIMORE, MARYLAND 21218

PROJECT TITLE: Ferromagnetic Relaxation in Ultrathin Films  
of Amorphous Alloys

PROJECT DIRECTOR: PRINCIPAL INVESTIGATOR (GEORGE T. RADO)

NAME: George T. Rado  
TITLE: Research Professor of Physics  
SOC. SEC. NO.: 577-60-6192

TYPE OF APPLICATION: ☒ NEW PROJECT  
☐ CONTINUATION  
☐ RENEWAL  
☐ SUPPLEMENT TO  
☐ REVISION TO PROPOSAL

AMOUNT REQUESTED FROM SPONSOR: \$37,000

PROPOSED STARTING DATE: May 1, 1985

PROPOSED DURATION IN MONTHS: 7

ENDORSEMENTS: PROJECT DIRECTOR (CORRESPONDENT) DEPARTMENT CHAIRMAN

SIGNATURE

NAME  
DIVISION  
TELEPHONE  
DATE

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22 March 1985

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ADMINISTRATIVE OFFICE

*John Dearden*  
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RESEARCH ADMINISTRATION  
COORDINATING OFFICE  
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DATE: \_\_\_\_\_

## Introduction

It has long been known that in devices designed for electromagnetic pulse (EMP) mitigation, the use of ferromagnetic rather than nonferromagnetic materials offers several advantages. Quite recently it was suggested that the shielding effectiveness of such devices might be further improved if their ferromagnetic components were made of amorphous rather than crystalline materials. The reason is that certain amorphous ferromagnets are highly flexible and that because of their unusually low magnetostriction the dependence of their magnetic properties on external stresses is relatively small.

## Statement of Work

Since the ferromagnetic relaxation behaviour of an amorphous (or crystalline) ferromagnetic material determines its magnetic losses, we believe that relaxation is among the most important properties which govern the effectiveness of a ferromagnetic material for EMP shielding. It appears, however, that the literature contains only a few experimental results and almost no theoretical insight on ferromagnetic relaxation in amorphous ferromagnets.

In the proposed research we plan to conduct experimental and theoretical investigations of ferromagnetic relaxation in amorphous materials. These materials will usually be in the form of ultrathin films whose thickness is about 200Å or less. The reason for concentrating our studies on ultrathin films is twofold: Firstly, such films have been entirely omitted in previous investigations of ferromagnetic relaxation in amorphous materials, and secondly, preliminary experimental results by the present Principal Investigator (P.I.) strongly indicate that in at least one amorphous material the ferromagnetic resonance linewidth (which is related to ferromagnetic relaxation) of ultrathin films has a particularly interesting dependence on film thickness. We believe that these results are not only scientifically interesting but potentially useful for EMP shielding.

In addition to measuring the ferromagnetic resonance linewidth of ultrathin films, we plan to investigate various structures consisting of several alternating layers of ultrathin amorphous ferromagnets and highly conductive metals. With this flexible arrangement, we should be able to control independently the magnetic and conductive properties of layered structures and to make use of the interesting relaxation properties of ultrathin films for achieving a sufficiently large total shielding in a relatively small space.



Since our general goal is to obtain an increased knowledge of ferromagnetic relaxation in amorphous materials, we might find it necessary to measure spin wave linewidths in addition to ferromagnetic resonance linewidths. As to the specific materials to be investigated, our initial studies will involve various amorphous materials which are sufficiently simple so that there is some hope that their linewidths will be understandable. In subsequent studies, of course, we shall investigate amorphous materials having unusually low magnetostriction because the attainability of this property is clearly a necessary (although not sufficient) condition for optimizing EMP mitigation.

#### Deliverables

Monthly memorandum reports will be made to the NRL contract officer's scientific representative. When applicable, scientific journal articles on the work will be submitted for publication.

Budget For 7 Months

NOTE: The numbers which follow may be adjusted slightly but the total of \$37,000 will not be changed.

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Salaries and fringe benefits (explained below*)	\$15,800
Travel	1,500
Materials and supplies	4,200
Publication costs	1,500
Subtotal	<u>23,000</u>
Overhead (61% of Subtotal)	14,000
<u>Total</u>	<u>37,000</u>

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\*Explanation of salaries and fringe benefits:

A. Partial salary of Principal Investigator for 1 year	\$15,625
Fringe benefit on A (8.5%)	1,328
B. Partial salary of secretary for 1 year	2,000
Fringe benefit on B (25%)	500
C. Salary of graduate student for 1 year	7,500
Fringe benefit on C (8% of salary for 3 summer months)	150
Total salaries and fringe benefits for 1 year	<u>27,103</u>
Pro-rated for 7 months	<u>15,810</u>

## NRL Statement of Work

### PHASE I (Seven month program)

#### TASK I - Identification of experimental conditions

- 1.1 Identify an amorphous alloy system and suitable ferromagnetic compositions.
- 1.2 Identify thicknesses of ultrathin films.
- 1.3 Identify ferromagnetic resonance configurations and frequencies.

#### TASK II - Theory

- 2.1 Develop a ferromagnetic resonance theory for amorphous alloys which incorporates the effect of film surfaces on the resonance frequency of ultrathin films.

#### TASK III - Experiments

- 3.1 Prepare ultrathin films of amorphous alloys (see 1.1 and 1.2).
- 3.2 Perform ferromagnetic resonance experiments (see 1.3).
- 3.3 Determine experimentally the dependence of ferromagnetic resonance linewidth on film thickness.

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